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Donor concentration dependence of the gain of a dye mixture laser

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Abstract. A dye mixture laser consisting of 7-diethylamino-4-methyl coumarin (donor) and fluorescein disodium salt (acceptor) dissolved in 2methoxyethanol was investigated under nitrogen laser pumping. Gain was measured with the acceptor concentration kept fixed 1.5×10^{-3}M and the donor concentration changed from 0.0 to 2.5×10^{-3}M for pump powers of 8.5 kW and 6.4 kW. An expression for gain based on radiative and Forster type energy transfer was derived. The linear dependence of experimentally measured gain on donor concentration was in agreement with the theory.

1. Introduction

Energy transfer dye lasers have been studied previously by various authors (Mooller *et al* 1971, Ahmed *et al* 1974, Dunning and Stokes 1972, Hilborn and Brayman 1974, Diens and Madden 1973, Urisu and Kajiyama 1976) However the dependence of gain on donor concentration has not been studied so far. From such a study the efficiency of energy transfer can be determined by measuring the rate of increase of acceptor gain with increasing donor concentration. In this paper we report on a study of laser action in a mixture of 7-diethylamino-4 methyl coumarin (DAMC) and fluorescein disodium salt (FDS) in 2 methoxyethanol The small signal gain was measured with the FDS concentration fixed at $1{\cdot}5\times10^{-3}\,M$ and the donor concentration changed from 0·0 to $2{\cdot}5\times10^{-3}\,M$ for two pump powers of 8·5 kW and 6·4 kW. A theoretical analysis of the system gave results which agreed well with the experimental findings.

2. Experimental

A nitrogen laser constructed in our laboratory with a repetition rate variable from 1 pps to 10 pps was used to transversely pump the dye solution in a quartz

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cuvette. Gain was calculated using the method of Shank *et al* (1970). The N_2 laser power was measured using a calibrated photodiode (ITL FD50) and was found to be stable to within 5%. The photodiode output was fed to a Tektronix (485) oscilloscope and the full width at reciprocal e (FWRE) of the N_2 laser output was found to be 12 nsec The dye laser output was monitored at 540 nm using an Andhra Scientific Co prism monochromator with an RCA-931A photomultiplier. The overall resolution was 2 nm. The gain per molecule was plotted against the fractional donor concentration with acceptor concentration fixed at $1{\cdot}5\times10^{-3}M$ Slopes of the gain vs donor concentration curves were calculated from a linear least square fit of the data points.

3. Theory and discussion

Here we assume that energy transfer for the DAMC-FDS system is due to radiative transfer and the Förster type nonradiative transfer 8,9 with rate constants k_R and k_F respectively. The rate equations for the mixture system can be written in a dimensionless form as

$$\frac{dn_{SD}}{dx} = n_{OD}\gamma_D - k_F N n_{SD} n_{DA} \tau_A - n_{SD}\frac{\tau_A}{\tau_D}$$

$$\frac{dn_{SA}}{dx} = n_{OA}\gamma_A + (k_F + k_R) N n_{OA} n_{SD} \tau_A - n_{SA}$$

where the subscript S indicates the first excited singlet state, O the ground state, A the acceptor and D the donor The n values are fractional state populations. The total population is $N = N_A + N_D$, the sum of total donor and acceptor populations $x = t/\tau_A$, where t is the elapsed time after start of pumping and τ_A the acceptor lifetime τ_D is the donor lifetime. γ_j = pump parameter = $\tau_j \sigma_j^L P(t)$ with $j = A$ or D where σ_j^L is the absorption cross-section at the N_2 laser wavelength and $P(t)$ the pump rate (photons cm^{-2} sec^{-1}) Under steady state conditions (Urisu and Kajiyama 1976) we can write

$$n_{SA} = F_A\gamma_A + \frac{(k_F + k_R) N_A F_D \gamma_D}{(1/\tau_D + k_F N_A)}$$

where $F_j = N_j/N$, and the small signal gain becomes

$$g(\lambda) = \left[\frac{(k_F + k_R) N_A \sigma_{SEA}(\lambda)\gamma_D}{(1/\tau_D + k_F N_A)} - \sigma_{SEA}(\lambda)\gamma_A + \sigma_{SAA}(\lambda)\right] F_D + \sigma_{SEA}(\lambda)\gamma_A - \sigma_{SAA}(\lambda)$$

where $g(\lambda)$ is the gain per molecule, $\sigma_{SEA}(\lambda)$ is the emission cross-section and $\sigma_{SAA}(\lambda)$ the absorption cross section at 540 nm for FDS The donor absorption and emission cross sections at 540 nm are negligibly small

From this equation one finds that the gain per molecule is proportional to the fractional donor concentration for a fixed acceptor concentration N_A.

This conclusion is borne out by experiment as is evident from Figure 1. For a given pump power and acceptor concentration the slope gives a measure of the efficiency of energy transfer for the dye mixture system. Thus this method can be used to find the best donor dye for a given acceptor dye and also quantitatively determine the transfer efficiency. In the case of DAMC, FDS dye mixture system the slopes are $7{\cdot}59\times10^{-18}$ cm^2 for 8·5 kW and $6{\cdot}89\times10^{-18}$ cm^2 for 6·4 kW, for $N_A = 1{\cdot}5\times10^{-3}$ M

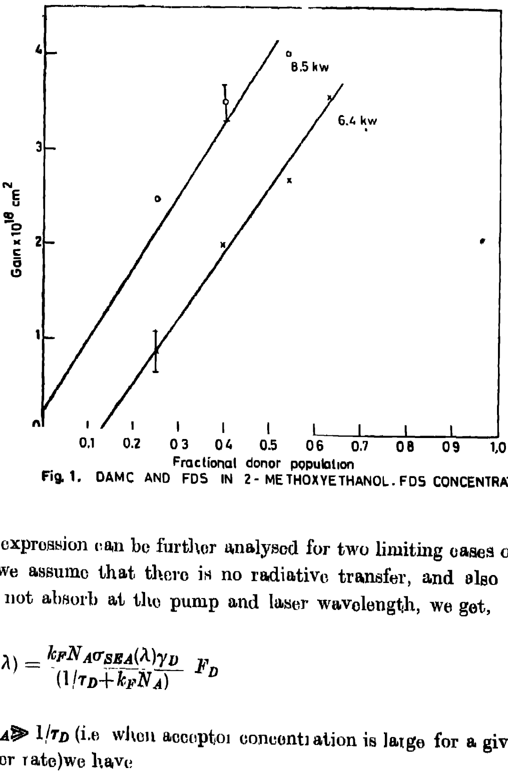

Fig. 1. DAMC AND FDS IN 2-METHOXYETHANOL. FDS CONCENTRATION 1.5x10^{-3} M

The gain expression can be further analysed for two limiting cases of energy transfer. If we assume that there is no radiative transfer, and also that the acceptor does not absorb at the pump and laser wavelength, we get,

$$g(\lambda) = \frac{k_F N_A \sigma_{SEA}(\lambda)\gamma_D}{(1/\tau_D + k_F N_A)} F_D$$

further if $k_F N_A \gg 1/\tau_D$ (i.e when acceptor concentration is large for a given large Forster transfer rate) we have

$$g_F = g(\lambda) = F_D \gamma_D \sigma_{SEA}(\lambda)$$

For the other extreme case of purely radiative transfer, with the condition that the acceptor does not absorb at the pump and laser wavelength, we have

$$g_R = g(\lambda) = k_R \tau_D N_A F_D \gamma_D \sigma_{SEA}(\lambda).$$

Thus if Forster type transfer predominates the gain varies as F_D and if radiative transfer predominates, the gain varies as $N_A F_D$. Hence by studying the gain as a function of donor as well as acceptor concentrations it should be possible to get an idea of the predominating energy transfer mechanisms. Further work in this line is being carried out by us and the results will be published elsewhere.

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